EUROPHYSICS LETTERS

Europhys. Lett., (), pp. ()

Reply to the comment of Chudnovsky&Garanin on "Spin relaxation in Mn_{12}-acetate"

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(received ; accepted)

PACS. 75.45.+j – Macroscopic quantum phenomena in magnetic systems.
PACS. 75.50.Xx – Molecular Magnets.
PACS. 75.30.Pd – Surface magnetism.

Angular momentum conservation is a physical law that must be obeyed in any closed system. However, it is well known from standard textbooks that in the case of interactions with phonons a part of the total momentum and of the total angular momentum can be absorbed by the whole crystal and is then no longer considered (cf. e.g. the Mössbauer effect[1] and the Umklapp process[2]). Therefore, there is no such general reason why the spin-phonon couplings

$$g_1(\epsilon_{xx} - \epsilon_{yy}) \otimes (S_x^2 - S_y^2) + \frac{1}{2} g_2 \epsilon_{xy} \otimes \{S_x, S_y\} \tag{1}$$

must vanish in the spin-phonon Hamiltonian $\mathcal{H}_{\rm sp}$ (see Eq. (6) in Ref. [7]) due to angular momentum conservation as incorrectly claimed in Ref. [3]. Both terms in Eq. (1) were used e.g. in Refs. [4], [5], [6], and [7] to describe phonon-assisted tunneling in Mn_{12}. Furthermore, such terms were derived in Refs. [8] and [9] many years ago by means of perturbation theory: These authors combined two spin-orbit interactions $\mathcal{H}_{\rm SO} = \lambda \mathbf{L} \cdot \mathbf{S}$ with one orbit-lattice interaction $\mathcal{H}_{\rm OL}$, which yields a third-order term of the form[8]

$$\sum_{j,k} \frac{\mathcal{H}_{ij}\mathcal{H}_{jk}\mathcal{H}_{ki'}}{\varepsilon_{ij}\varepsilon_{ik}}, \tag{2}$$

where $\varepsilon_{ij} = \varepsilon_i - \varepsilon_j$ are differences between the eigenenergies of the unperturbed Hamiltonian $\mathcal{H}_{\rm O} + \mathcal{H}_{\rm L}$. $\mathcal{H}_{\rm O}$ is the Hamiltonian of the free ion, and $\mathcal{H}_{\rm L}$ the Hamiltonian of the lattice. Note that the orbit-lattice interaction

$$\mathcal{H}_{\rm OL} = \sum_f \frac{\partial V}{\partial Q_f} Q_f + \frac{1}{2} \sum_{ff'} \frac{\partial^2 V}{\partial Q_f \partial Q_{f'}} Q_f Q_{f'} + \cdots, \tag{3}$$

where V is the energy due to the crystalline electric field, includes terms that are *linear in normal displacements* Q_f, which arise from the Jahn-Teller effect[8, 10, 11]. As a matter of fact, the Jahn-Teller effect is very strong in Mn_{12}[12]. In order to provide the connection between Eq. (3) and Eq. (1), we note that the strain components $\epsilon_{\alpha\beta}$, $\alpha, \beta = x, y, z$, are proportional to the Fourier transforms $u_{\mathbf{q}}$ of the displacements Q_f[4, 7], which can be represented by

phonon creation and annihilation operators with wave vectors $\mathbf{q}$, i.e. $u_{\mathbf{q}} \propto (a_{\mathbf{q}} + a^\dagger_{\mathbf{q}})$[4, 7, 9]. Obviously, Eqs. (2) and (3) imply Eq. (1) and that part of the angular momentum is absorbed by the whole lattice, which is as usual assumed to be infinitely heavy and of infinite moment of inertia.

Moreover, it was shown generally in Ref. [9] that the spin-phonon coupling constants corresponding to our g_i, $i = 1, 2, 3, 4$, are all of the same order of magnitude. This result was also obtained by Van Vleck[8] for Cr^{3+}.

We agree with Ref. [3] that the g_1 spin-phonon coupling term given in Eq. (3) of Ref. [6] and in Eq. (6) of Ref. [7] is not due to rigid rotations only, as first pointed out to us by J. Villain. However, we know that $g_4 = 2A$[4, 6, 7, 13], from which we obtain that $g_1 \approx A$[8, 9]. Thus, terms of the form (1) do exist in Mn_{12} in contrast to the incorrect claim made in Ref. [3].

Finally we conclude with the following erratum to Refs. [6] and [7]: The equation $g_1 = A$ (see Ref. [6] and Eq. (19) in Ref. [7]) must be changed to

$$g_1 \approx A. \tag{4}$$

Our fits shown in Refs. [6] and [7], which are in good agreement with experimental data, confirm Eq. (4). We emphasize that all our conclusions presented in Refs. [6] and [7] remain unaffected by this change.

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